

VAPORIZATION OF ELEMENTAL MERCURY FROM MOLTEN LEAD AT LOW CONCENTRATIONS

by

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ABSTRACT

Experiments were conducted to measure the rate of vaporization of elemental mercury from molten lead to provide a basis for estimating radiological source terms for the APT (Accelerator Production of Tritium project) lead blanket. These data also have application to other accelerator targets in which mercury may be created by proton spallation in lead. Molten pools of lead with from 0.01% to 0.10% mercury were prepared under inert conditions. Experiments were conducted which varied in duration from several hours to as long as a month to measure the mercury vaporization from the lead pools. The melt pools and gas atmospheres were controlled at 340°C during the tests, above the melting temperature of lead. Parameters which were varied in the tests included the mercury concentrations, gas flow rates over the melt, circulation in the melts, gas atmosphere compositions and the addition of aluminum to the melts. The vaporization of mercury was found to scale roughly linearly with the concentration of mercury in the pool. Variations in the gas flow rates were not found to have any effect on the mass transfer, however circulation of the melt by a submerged stirrer did enhance the mercury vaporization rate. The rate of mercury vaporization under a high-purity argon atmosphere was found to exceed that for an air atmosphere by as much as a factor of from ten to 20; the causal factor in this variation was the formation of an oxide layer over the melt pool with the air atmosphere which retarded mass transfer across the melt-atmosphere interface. Aluminum was introduced into the melt to investigate its effect upon the mercury vaporization rate. No effect was observed for a case under a high-purity argon atmosphere, which suggests that there are no chemical effects of the aluminum on the vaporization kinetics. With an air atmosphere, the presence of aluminum in the melt reduced the mercury vaporization by a factor of six in comparison to the identical test but without aluminum, suggesting that aluminum in the lead/mercury melt retards the vaporization of mercury by creating a surface oxide layer in addition to the lead-oxide layer or by changing the character of the lead-oxide layer, thereby increasing the mass transfer resistance.

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1. INTRODUCTION

The primary contributors to the radiological source term for APT come from the proton-irradiated tungsten spallation target and the surrounding lead/aluminum blanket. From the tungsten spallation target, the principal spallation products that are considered in accident analyses are activated isotopes of tungsten. From the lead blanket, the principal spallation products considered are isotopes of mercury. The present investigation attempts to measure the release of elemental mercury from molten lead to provide data for estimating accidental mercury releases as a function of several dominant parameters such as temperature, mercury concentration and mass transfer enhancement. Isotopes of mercury would be produced in the lead blanket as a result of nuclear reactions in the lead with protons and neutrons from the tungsten target. In the highest power regions of the lead blanket, the concentration of mercury could approach 0.012% after one year of operation at full accelerator power (170 MW) and availability (75%). The mercury concentration could approach 0.06% if these modules were in service for five years [1-10].

Should coolant be lost in the target system and the decay heat in the target be deposited in the surrounding lead by thermal radiation, temperatures in the lead could exceed structural limits and cause a physical collapse of the blanket modules into a non-coolable geometry before melting. Such a sequence of unmitigated events could result in some melting of the lead blanket and create the potential for the release of mercury from the damaged modules into the target-blanket confinement. The conditions which could be expected would be temperatures close to the lead melting temperature, an air or air/steam atmosphere, upper-limit mercury concentrations of 0.012%, and aluminum in the melt from the blanket structures. The lead in this study was maintained above melting for practical reasons. The series of experiments which will be discussed provide an empirical basis from which mercury release estimates may be made and parametric variations considered.

2. EXPERIMENTAL APPARATUS

The apparatus used in this study was designed and constructed to incorporate features that enhance its safety, reliability, and ease of use. The entire apparatus was constructed on a lab rack in a stainless steel secondary containment pan mounted on a test stand in a large lab fume hood. The pan provided protection from potential spills or leaks of mercury, and the fume hood provided protection from mercury vapors in the event of the failure of a cold trap. The apparatus was constructed from commercial hardware and labware incorporating standard tapered joint connections for the glass components, facilitating fast servicing. A schematic of the test apparatus is shown in Fig. 1.

The melt vessel was assembled using a 316 stainless steel hemispherical reaction flask with a four-inch neck opening and a Pyrex three-neck reaction flask head. Both pieces were mounted to the test stand as a single unit using a quick release design clamp and flat Teflon gasket. A heavy-duty hemispherical heating mantle was mounted immediately below the reaction flask. This heating mantle was equipped with several type-K thermocouples, a temperature controller, and a supplemental insulation package to provide stable control of the melt temperature. The left-hand neck of the flask top was connected to a gas supply system capable of delivering heated air or high-purity pre-purified argon (99.998%) to the vessel via mass flow controllers. The center neck of the flask top was equipped with a Trubore stirrer assembly driven by a remotely mounted variable speed motor via a flexible shaft. The stirrer blade is a chordal segment of a circle with radius 1.75 inches and a chord length of 2.792 inches. The right-hand neck of the flask top was fitted with a Pyrex transfer line leading to the cold trap. This

transfer line, shaped like an inverted U, provided approximately eight inches of lateral separation between the reaction flask and the cold trap. To prevent premature condensation of mercury vapors, this line was trace heated with a heating tape and equipped with type-K thermocouples and a temperature controller to maintain a wall temperature of about 100°C. A series of U shaped cold traps were fabricated from Pyrex to be installed as required on the end of the transfer line. A Dewar flask containing a slurry of dry ice and acetone was raised up around the cold traps after they were installed on the transfer line to cause the mercury vapors in the flowing gas stream to condense. All glass joints were sealed with standard Teflon tapered joint sleeves. All glass surfaces and gas supply lines were either heavily insulated or trace heated to prevent accidental mercury condensation.

3. EXPERIMENTAL PROCEDURES

The following procedure was employed to conduct the mercury vaporization tests. An initial charge of about 2.5 kg of high-purity lead strip was accurately weighed on a Sartorius electronic top loading balance to a precision of 0.1 gram. The reaction flask was initially heated above the lead melting temperature to approximately 350°C and a high-purity argon cover gas flow was established through the apparatus. The stirrer assembly normally installed in the center neck of the flask was removed and the center neck used to introduce the lead to the flask. Lead strips were continuously added to the flask as they melted into the pool. When all of the lead was melted into the pool, it was stirred manually and skimmed of slag. This slag was collected and weighed and the mass of slag subtracted from the initial mass of lead to determine the initial mass of the pool. High-purity argon was used exclusively to avoid introducing gaseous impurities into the reaction flask. On the one occasion in which low-purity argon was inadvertently supplied, the results were obvious and the experiment was abandoned. Quality control measures were then instituted to avoid this problem. The temperature of the pool as well as the temperature of the cover gas and transfer line were adjusted and stabilized at the desired set points. The stirrer assembly was then installed in the center neck and set to stir the pool in preparation for mercury injection.

A hypodermic syringe with a long spinal needle was used to inject the mercury into the pool. The syringe and needle were tare weighed using a Sartorius analytical balance capable of weighing to a precision of 0.1 mg. The balance calibration was frequently checked using both its internal 100 g standard mass and an external 50 g standard mass. An amount of mercury appropriate to give the desired mercury concentration was taken up in the syringe and the syringe reweighed. The transfer line was temporarily disconnected from the flask and, with vigorous stirring, the mercury was injected into the bulk of the pool to achieve the desired mercury concentration. Post-injection mass measurement of the syringe was recorded to calculate the actual mercury concentration. The transfer line was then reconnected and a tare-weighed set-up collection trap installed on the end. A Dewar flask containing a slurry of crushed dry ice and acetone was elevated around the sample collection trap. This slurry maintained the trap at about -80°C, a temperature at which mercury has virtually no vapor pressure [11]. Stirring, gas species temperature and flow rate, pool temperature, dry ice replacement and trap change outs were attended to as prescribed in run sheets prepared for each test. The lead and gas atmosphere temperatures were measured and controlled with standard type-K thermocouples, each with an uncertainty of 2.5°C. The gas flows were calibrated by using a “soap bubble flow meter” to time the delivery of a fixed volume of gas. The flow meter is constructed from a standard 100 ml laboratory burette which has a volume tolerance of 0.2 ml. For the 200 ml/min flow setting, the uncertainty in the calibration is estimated to be 6%; for the 750 ml/min setting, the

uncertainty in the calibration is estimated to be 2%. As traps were changed out, they were placed in a desiccator prior to weighing to remove atmospheric moisture that condensed on their cold (-80°C) surfaces. The uncertainty in elapsed time was negligible. When dry, the traps were weighed and their masses compared to pre-run masses to determine the mass of mercury collected. The masses of mercury injected into the melt as well as the masses of mercury collected in the cold traps were measured to an accuracy of ± 0.5 mg.

4. CONDUCT OF EXPERIMENTS

A series of tests was executed to quantify the effects of several dominant parameters upon the rate of vaporization of elemental mercury from a pool of molten lead. The matrix of parameters which was considered for these tests included variations in melt composition (Pb, Pb/Hg), gas composition (air, high-purity argon), gas flow rates (low/high), aluminum in the melt and circulation of the melt. The experimental ranges of these parameters are listed in Table 1. A discussion of each of the tests and their results is given below. The reader will note that there is no test designated as Hg-Pb/07. This test failed during execution and the run number was not reused. Several tables which summarize the test parameters and results have been provided for reference. Table 1 provides a listing of the nominal test parameters and their ranges. The specifications for the individual tests are provided in Table 2. Table 3 provides a comprehensive listing of the experimental results of the ten tests. Tables 4-8 provide the detailed mercury release measurements for each test.

4.1 Experiment Hg-Pb/00: Pb only, argon

This test was the first in the series and was conducted primarily to provide a shakedown test of the experimental apparatus. All the features of the apparatus were exercised including the melt, gas, and trace heaters, as well as the dry-ice/acetone cold trap for condensation and collection of mercury vapors. The heating systems achieved and maintained pre-established set points after several modifications to the heater configuration and insulation packaging. A second objective of the test was to address an issue which was raised during review, and this was the possibility of lead being carried over from the melt crucible into the heated gas-discharge plumbing and deposited in the cold traps, thus affecting the mercury mass measurements. The concern was whether the mass collected in the cold traps would be all mercury or would it be a lead/mercury mixture requiring complex post-experiment chemical analyses to sort out. Although the data for this experiment were null and are not reported in Table 3, the data from this experiment demonstrated that there was no measurable increase in the mass of the cold traps after more than 24 hours at the prescribed test conditions of 340°C for both melt and flowing high-purity argon, thus there was no lead collected in the cold trap. These results confirmed assumptions during the design phase that only volatile components in the melt would accumulate in the cold traps and that, if high-purity lead sheet material were used for the melt preparation, the collection of volatile contaminants could be eliminated.

4.2 Experiment Hg-Pb/01: 1.0% Hg, argon

The second test in the series, designated as Hg-Pb/01, was conducted to evaluate mercury injection and mixing techniques, the cold trap collection efficiency, and initial mercury vaporization results for a case with a bounding concentration, in this case 1% mercury by weight. This experiment helped to finalize procedures and techniques for the mercury injection and mixing. The dry ice/acetone cold trap technique for condensing and collecting the mercury vapors which were released from the molten pool worked as intended. All the mercury was found to condense in the U-tube at the dry ice/acetone

interface in small droplets on the inside of the glass tube. Since no mercury vapor was able to be detected at the discharge of the U-tube, it was concluded that the cold trap technique was 100% efficient at condensing and collecting mercury vapors. At the outset, vaporization of mercury from the molten lead pool at this high concentration was immediate and excessive. Greyish deposits of mercury formed on all the glass surfaces immediately and, over a period of two hours, nearly 5% of the mercury in the lead was collected in the cold trap. The average mercury release, extrapolated to a 24-hour period, was calculated to be 56% per day. The test was terminated and it was decided to restrict the mercury concentration in the lead to a maximum of 0.1% for future tests, with some tests conducted at 0.01% to evaluate the scaling of the mercury vaporization rate with the mercury concentration.

4.3 Experiment Hg-Pb/02a, b: 0.1% Hg, argon

The first quantitative test in the series was Hg-Pb/02. The parameters of this test were as follows: lead pool 2.5 kg (nominally identical for all tests), mercury concentration 0.11% and a high-purity argon cover gas. There was no melt circulation during the test, gas flow was low (200 ml/min) and there was no aluminum added to the melt. This test was inadvertently performed in two stages. Hg-Pb/02a consisted of three sequential cold trap changes over an elapsed time of eight hours. The power to the melt heater was turned off after this elapsed time and the melt was allowed to solidify. The three cold traps were desiccated to remove all moisture as was the procedure with all cold traps and then weighed to determine the cumulative mass release of mercury over the eight hour period. It was observed from the results that the mercury release was still continuing after eight hours; as a result, the test was resumed with the designation Hg-Pb/02b, using the same melt charge and the test was run out to a total duration of 48 hours. The combined results of Hg-Pb/02a and Hg-Pb/02b are shown in Fig. 2. The total mass of mercury released after 48 hours was found to be 0.33 g out of an initial inventory of 2.83 g, a release fraction of 12% or 5.9% per day on average. The results of this test are listed in Table 4. [Note: In a later comparison of results, Hg-Pb/02 will be superseded by Hg-Pb/11 which was a repeat of Hg-Pb/02 but was uninterrupted over a period of seven days].

4.4 Experiment Hg-Pb/03: 0.01% Hg, argon

The parameters for this test were identical to those for the previous test with the exception of the mercury concentration. The mercury concentration for this test was reduced by a factor of ten to 0.012% to provide a comparison to the results of Hg-Pb/02 on the basis of initial mercury concentration. All other parameters were unchanged. The mercury concentration of 0.01% is nearly equal to the maximum calculated mercury concentration in the APT blanket after one year of operation. This test, building on the lessons from the previous test, was set up to run for seven days. The results of this test are shown in Fig. 2 along with the previous test (Hg-Pb/02). The total fraction of mercury released from the melt was found to be 10.2% over a period of nearly seven days. This computes to an average release fraction per day of 1.5%. The average mass flux per unit area is 2.2×10^{-6} g/cm² hr. However, it is more reliable to compare the results to Hg-Pb/11 instead of Hg-Pb/02 for reasons already discussed. The average mass flux per area for Hg-Pb/11 was 4.2×10^{-5} g/cm² hr. This comparison shows that the release rate scales nearly linearly with concentration. The concentration ratio for Hg-Pb/03 to Hg-Pb/11 is about 8.6 while the vaporization mass flux/unit area ratio is about 19.3, about a factor of two difference which is reasonable. The results of this test are listed in Table 4.

4.5 Experiment Hg-Pb/04: 0.1% Hg, argon, high gas flow rate

The purpose of this test was to examine the effect of mass-transfer resistance of the gas flow rate on the rate of vaporization of mercury from molten pools of lead. The mercury concentration was established at 0.11%, the gas and melt temperatures were set to 340°C as in all the other tests, and the test was begun with the nominal low gas flow rate of 200 ml/min. After 26 hours (1583 minutes), the gas flow was increased to 750 ml/min, and after 43 hours (2588 minutes) the gas flow was reduced back to 200 ml/min. The data are shown in Fig. 3 along with the data for Hg-Pb/05 (which examined the effects of circulation in the melt). It is clear from Fig. 3 that although the gas flow was increased by almost a factor of four in the middle of the test, there is no apparent change in the rate of mercury vaporization from the melt. It was concluded from this test that gas-phase mass transfer is not a significant resistance to the vaporization rate and, thus, not a significant parameter to consider. The fractional mercury release per day was measured to be 2.7%, comparable to tests Hg-Pb/02, 03, 10 and 11. The results of this test are listed in Table 5.

4.6 Experiment Hg-Pb/05: 0.1% Hg, argon, melt circulation

The purpose of this test was to examine the effect of circulation in the melt pool on the rate of vaporization of mercury from a molten pool of lead. The mercury concentration was established at 0.11% and the gas and melt temperatures were held at 340°C. The cover gas was high-purity argon and the flow was set to 200 ml/min. The glass stirrer which was used to initially mix the mercury in the melt was left submerged during the experiment. For the approximate time period 1200 to 2700 minutes (24.5 hours), the stirrer was turned on in the melt at a rate of approximately one rpm. After 2700 minutes, the stirrer was once again turned off. The data are shown in Fig. 3 along with the data for Hg-Pb/04. The vaporization rate began at a low rate but was observed to suddenly increase until, at about 2700 minutes, the release rate once again flattened to a low value. The mercury release fraction during the melt stirring period was found to be nearly 21% per day. For the times prior to stirring and after stirring, the normalized fractional mercury releases were 0.25% per day and 2.1% per day, respectively. It should be noted that this effect was only investigated at one concentration, 0.1% mercury. It was concluded that circulation in the lead pool would significantly increase the mercury vaporization rate above the stagnant case by replenishing the mercury in the surface layer which had vaporized. The results of this test are listed in Table 5.

4.7 Experiment Hg-Pb/06: 0.1% Hg, air

To examine the effect of an oxidizing atmosphere instead of an inert atmosphere, the following set of experiments (Hg-Pb/06-09) were conducted with air. It was not possible to use steam because of the cold traps. The first test in this set was Hg-Pb/06, which had the default set of test parameters (200 ml/min gas flow, no melt agitation, 0.10% mercury concentration) with the exception that an air atmosphere was used instead of high-purity argon. Melt and gas temperatures were set to 340°C as usual. The rate of mercury release was relatively high at the beginning of the test for the first collection period of 15 hours. After the initial collection period, the cold traps were replaced on 24-hour intervals to a total elapsed time for the test of 14 days. After the first mass collection, the collected mass decreased over the remainder of the 14 days by a factor of from 10 to 50, due to a lead oxide film which was observed to form on the surface of the pool. Whereas the mass collected in the first 24-hour period was 86 mg, in the second 24-hour period only 5 mg were collected and, in the 14th day, less than 2 mg was collected. The results of this test are shown in Figs. 4 and 5. Whereas for the earlier tests under a high-purity argon atmosphere the average daily mercury release fractions

were in the range 3% to 6% per day (for an initial mercury concentration of 0.1%), for Hg-Pb/06 the average daily release fraction was only 0.3%, a reduction in the vaporization rate by a factor of 10-20.

4.8 Experiment Hg-Pb/08: 0.01% Hg, air

This test was conducted to reproduce the conditions of test Hg-Pb/06 with the one exception that the mercury concentration was reduced to 0.011% to examine the effect of the Hg concentration in the presence of an oxidizing (air) atmosphere. The overall duration of this test was increased from 14 days to 27 days in the expectation of a greatly reduced Hg release rate. The mass of mercury collected in the first 48 hour period was only 2.4 mg, compared to 86 mg of mercury collected in the first 24 hours of Hg-Pb/06. Only 8.5 mg more were collected over the remaining 25 days, less than 0.4 mg/day on average. At the end of 27 days, a total of 10.9 mg of mercury had been collected during this test and the mercury release rate had decreased essentially to zero. This compared to a total of 112 mg released in Hg-Pb/06, a factor of ten more at a mercury concentration a factor of ten more than this test. The results of Hg-Pb/08 are shown in Fig. 4 along with the results of Hg-Pb/06 for comparison. The conclusions from this comparison are that the mercury release rate does scale with mercury concentration and the formation of a surface oxide layer due to an oxidizing atmosphere presents a significant resistance to vaporization. The results of this test are listed in Table 6.

4.9 Experiment Hg-Pb/09: 0.1% Hg, air, aluminum in melt

This test was a repeat of all the test parameters from a previous test (Hg-Pb/06: air atmosphere and 0.1% mercury) with the addition of a new test parameter, the addition of aluminum to the molten lead pool. Although the solubility of aluminum in lead at 340°C is extremely slight (only 0.04% at 500°C), aluminum can have a strong effect upon chemical reactions by altering the oxygen potential of the melt. To introduce aluminum into the melt, the glass stirrer which was used to mix the mercury was replaced by one made out of aluminum. To allow time for aluminum to dissolve into the melt from the stirrer paddle, this stirring period was increased from two hours to more than six hours; the aluminum stirrer was then left submerged in the melt for the duration of the test to allow for continued dissolution. Hg-Pb/09 was run for a total duration of 14 days. Gravimetric analyses of the cold traps indicated that out of a total mercury release of 16.5 mg in 14 days, 5.7 mg was collected in the first 16 hours; the remaining 11 mg was released at a continuously decreasing rate for the duration of the 14-day test. The overall mercury release fraction for this test was 0.7% at a daily average release rate of 0.05% per day. These results are a factor of six less than the results from Hg-Pb/06, indicating a retarding effect of aluminum in the melt on the mercury vaporization rate with an air (oxidizing) atmosphere. These results are shown in Fig. 5 and listed in Table 7.

4.10 Experiment Hg-Pb/10: 0.1% Hg, argon, aluminum in melt

The hypothesis from Hg-Pb/09 was that an aluminum oxide film might form over the surface of the lead pool with aluminum in the melt and an air atmosphere, or that the aluminum altered the characteristics of the lead oxide film that was observed to form with air. In order to test this hypothesis, Hg-Pb/10 was conducted to repeat the conditions of Hg-Pb/09 but under a high-purity argon atmosphere instead of air, thus eliminating oxidation from consideration. The melt preparation procedures for Hg-Pb/10 were identical to Hg-Pb/09; however, after the mercury mixing and melt stirring period (at the start of the test), the gas flow was left on high-purity argon instead of changing to air. The test was conducted for seven days for direct comparison to Hg-Pb/11. Gravimetric analysis of the cold trap masses revealed a surprisingly close agreement between the two tests, with

many of the data points lying over one another as shown in Fig. 5. The pool surface was observed to remain shiny for the entire test, indicating that there was no oxidation of either the lead or aluminum. The masses and release fractions for Hg-Pb/10 and Hg-Pb/11 were nominally identical. The conclusion that is drawn from this comparison is that there is no effect of dissolved aluminum in a pool of molten lead (at 340°C) on the vaporization of mercury from the pool without an oxidizing atmosphere. However, with an oxidizing atmosphere, the effect of aluminum in the melt is to retard the vaporization; no enhancing effect of aluminum on the vaporization of mercury from the melt was found. The results of this test are listed in Table 8.

4.11 Experiment Hg-Pb/11: 0.1% Hg, argon - repeat of Hg-Pb/02

The last test in the series was Hg-Pb/11 which has been previously discussed out of order because it was intended to use these results to replace Hg-Pb/02 for reasons already discussed. This test was conducted under better experimental control than Hg-Pb/02 and ran smoothly for seven days. The details will not be repeated here; the parameters are listed in Table 3 and the results are shown in Fig. 5 and listed in Table 8. It is clear from Fig. 5 that Hg-Pb/11 and Hg-Pb/10 are in excellent agreement, supporting the earlier stated conclusions.

5. DISCUSSION OF RESULTS

Experiments were conducted to measure the vaporization of mercury from pools of molten lead with mixtures of lead and mercury, on the order of 0.01% to 0.10% by weight, to cover the range of mercury concentrations expected in the APT blanket. Melt and atmosphere temperatures were controlled to 340°C to represent the low melt temperatures of interest. Although the results would probably be different at higher temperatures, this study was limited to a temperature just above the lead melting temperature because it was felt that accident conditions could not support higher temperatures. It was found that the vaporization rates of mercury from a molten lead pool scaled roughly linearly with the mercury concentration. In other words, a reduction in the mercury concentration by a factor of ten resulted in a reduction in the mercury vaporization rate by about a factor of ten. This was found to be the case with both high-purity argon and air atmospheres over the melt pool. The rates of mercury release were found to be insensitive to changes in the gas flow rates, indicating that the limiting resistance to mass transfer was on the condensed-phase side of the melt-atmosphere interface, not in the gas phase. A subsequent test in which the melt was circulated by a stirrer confirmed this observation; the stirring at one rpm increased the mercury vaporization rate by approximately a factor of ten over the identical case without circulating the melt. This result is fortunate because it indicates that it is not necessary to consider complex flow fields in the gas because the vaporization rate is insensitive to the gas-phase mass transfer coefficient. It appears that there is little resistance to mass transfer in the gas even at the lowest gas flow rate tested. A high-purity argon atmosphere was always used to prepare the melts and maintained while the prescribed test conditions were being established. For the early tests (up to Hg-Pb/05), high-purity argon was used for the atmosphere over the melt during the test in order to avoid the complications that might occur with air, while investigating the parametric effects of mercury concentration and mass transfer on the mercury vaporization rates. However, for tests Hg-Pb/06 through Hg-Pb/09, air was used for the atmosphere over the molten lead pool to examine its effect. The use of air over the melt instead of high-purity argon demonstrated an order of magnitude reduction in the mercury vaporization rate, attributed to the formation of a lead oxide layer over the melt which retarded vaporization through the interface. Subsequent tests which had a lower mercury concentration with air demonstrated a

corresponding reduction in the mercury vaporization rate. The addition of aluminum to the melt was investigated to determine its effects under both high-purity argon and air atmospheres. Although no effect of aluminum in the melt on the mercury vaporization rate was observed in the test under high-purity argon (Hg-Pb/10) as compared to its counterpart test Hg-Pb/11 (w/o Al), a significant reduction in the mercury vaporization was observed in the identical test with air and aluminum in the melt (Hg-Pb/09) as compared to its counterpart test Hg-Pb/06 (w/o Al). It was obvious to the eye that a lead-oxide layer formed on the melt surface in the tests which used air for the cover gas over the molten lead. It is not obvious, however, that an aluminum-oxide layer was the cause for the reduced vaporization rates which were measured in test Hg-Pb/09. Although the effect is clear, the causal factor for the observed effect of aluminum has not been conclusively established.

6. CONCLUSIONS

The integrated release of elemental mercury from pools of molten lead was measured under a variety of fixed and variable parameters, including mercury concentration, lead pool and atmosphere temperatures, atmosphere composition and aluminum in the melt. In addition, variations in the rates of mass transfer in the gas and melt were investigated. A total of eleven tests were conducted. The following results were obtained from these experiments.

1. Mercury vaporization rates were measured to scale roughly linearly with the initial mercury concentration for concentrations in the range 0.01% to 0.10% by weight.
2. Variations in the gas flow rates across the surface of the molten lead pool had no measurable effect on the mercury vaporization rate, indicating that the mass transfer resistance in the gas could be neglected.
3. Variations in the melt pool agitation did have a significant effect upon the mercury vaporization rate, indicating that experimenters should take great care not to arbitrarily enhance the mercury vaporization rate by agitation of the melt.
4. Experiments in which air was used for the atmosphere over the melt instead of high-purity argon demonstrated an order-of-magnitude reduction in the mercury vaporization rate due to the formation of an oxide film on the lead surface which did not form when high-purity argon was used.
5. The addition of aluminum to the melt was examined for its effect on the mercury vaporization rate. The presence of aluminum in the melt had no apparent effect on the mercury vaporization rates in experiments under a high-purity argon atmosphere; however, a significant reduction in the mercury vaporization rate was measured in experiments with an air atmosphere with the addition of aluminum to the melt compared to the identical experiment with air but without aluminum in the melt.

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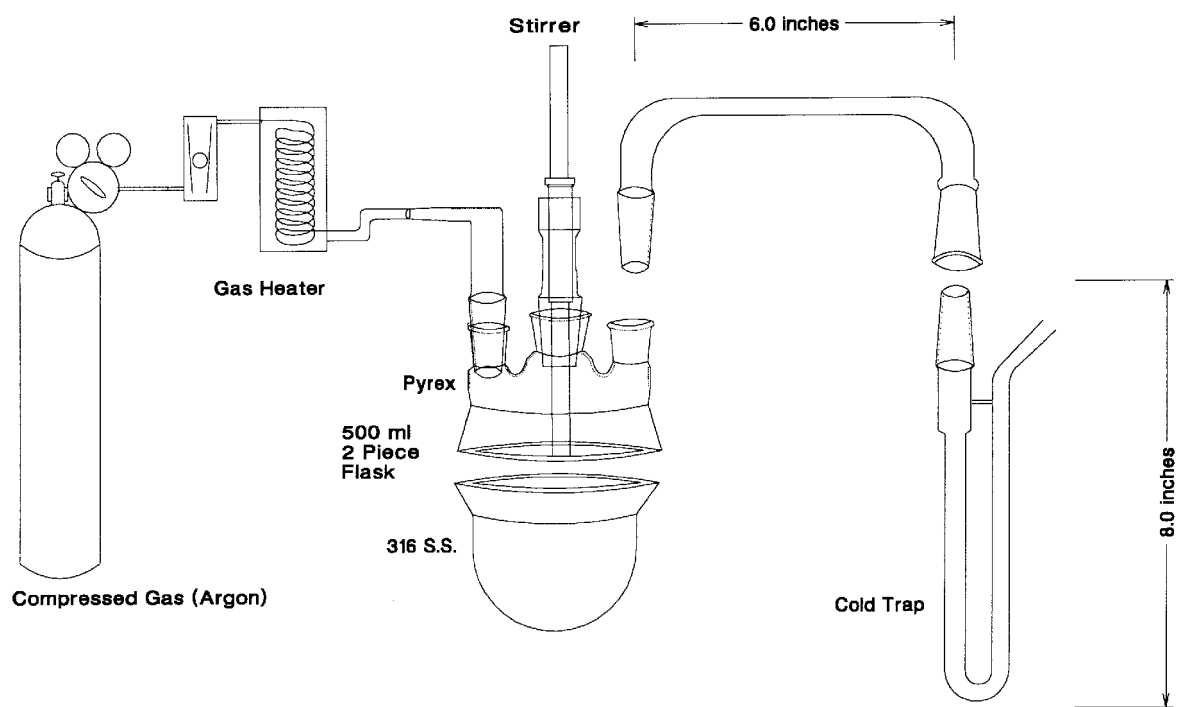


Figure 1. Schematic of the Experimental Test Facility.

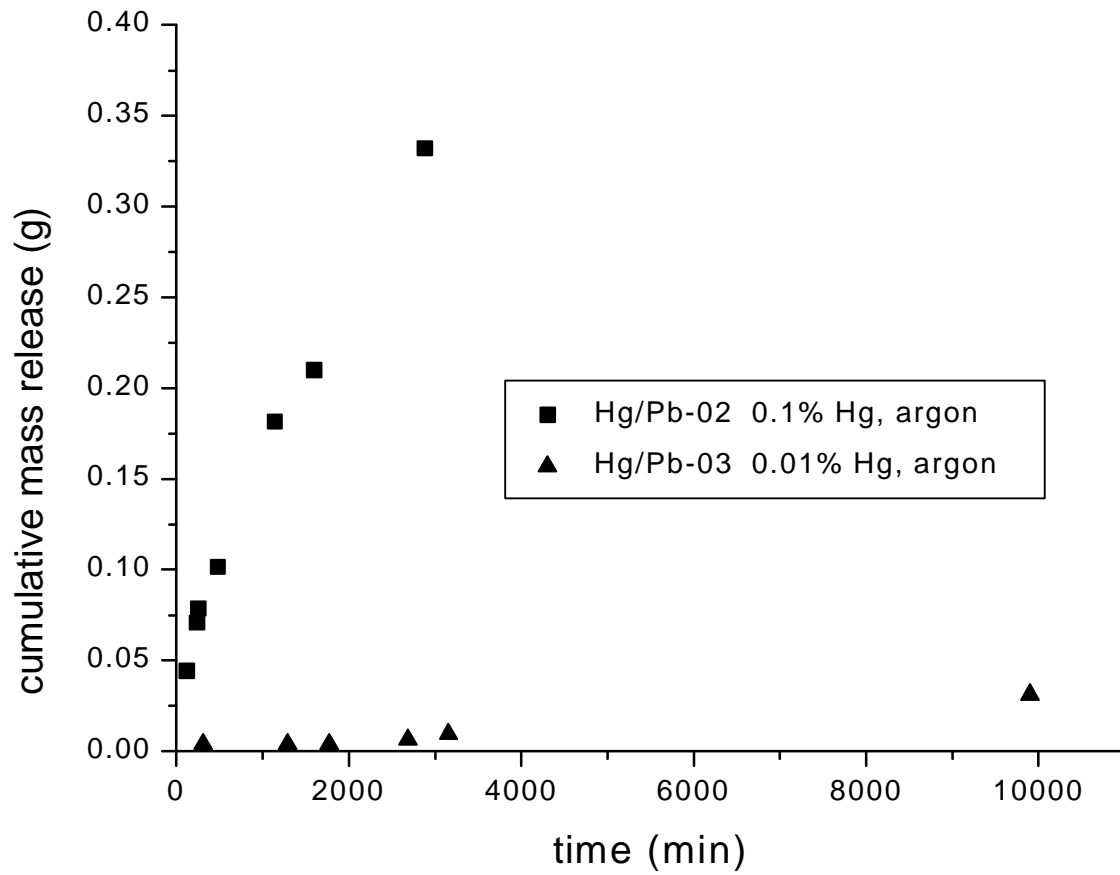


Figure 2. Cumulative Mercury Release From a Molten Lead Pool as a Function of Mercury Concentration under a High-Purity Argon Atmosphere: Hg-Pb/02 vs. Hg-Pb/03.

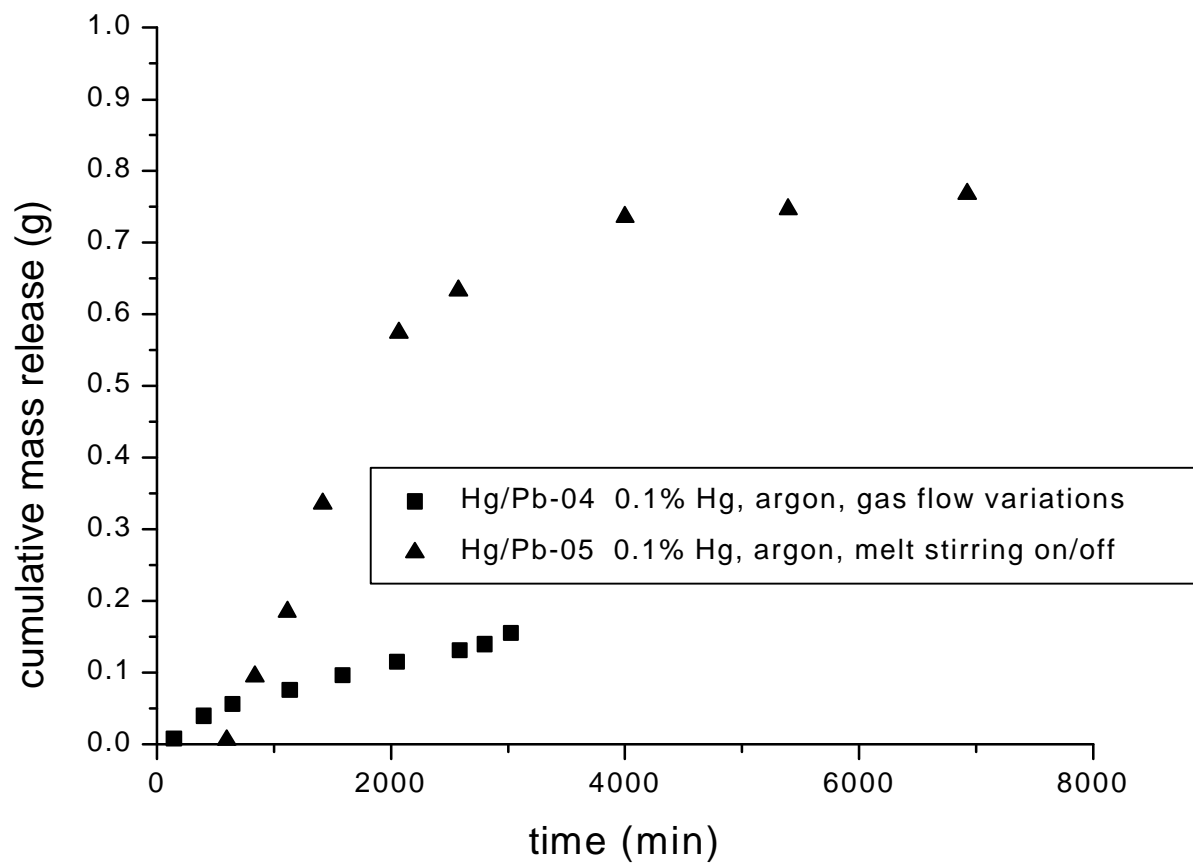


Figure 3. Cumulative Mercury Release From a Molten Lead Pool as a Function of Gas Phase and Condensed Phase Mass Transfer Enhancements: Hg-Pb/04 vs. Hg-Pb/05.

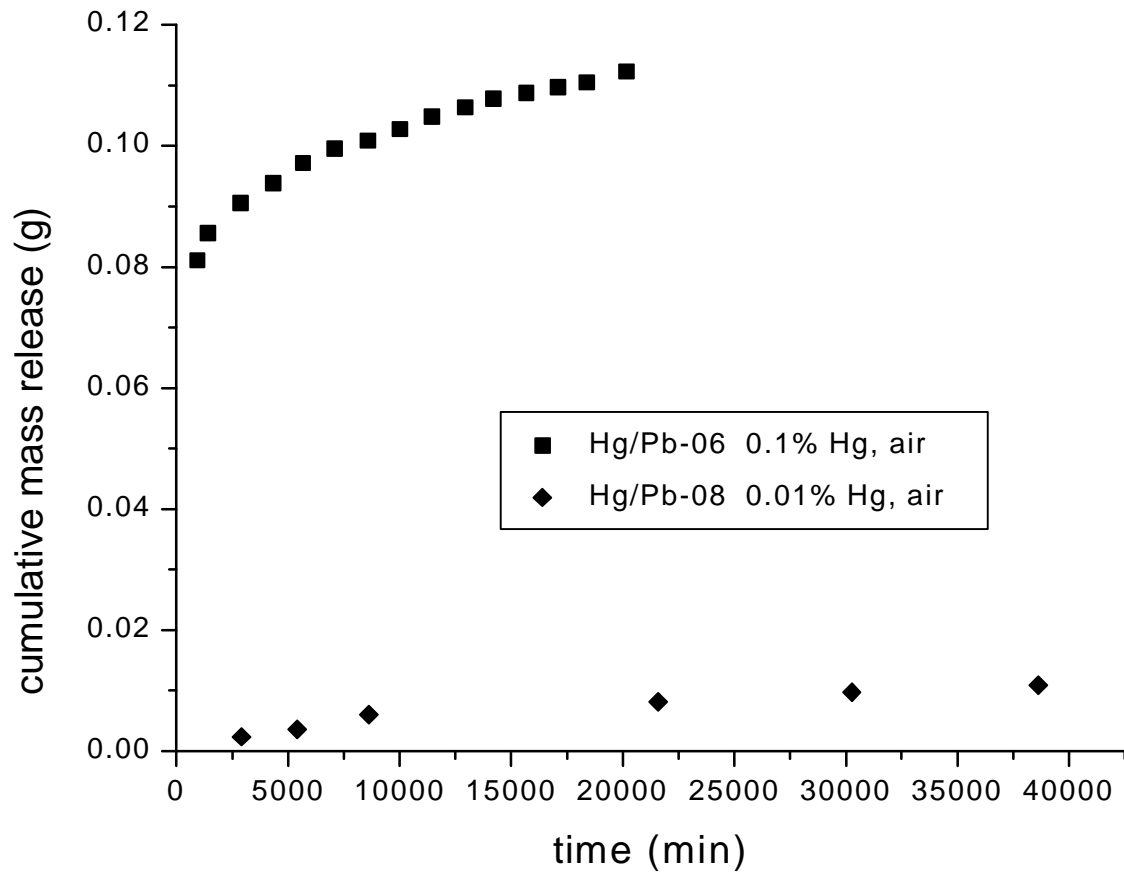


Figure 4. Cumulative Mercury Release From a Molten Lead Pool as a Function of Mercury Concentration with an Air Atmosphere: Hg-Pb/06 vs. Hg-Pb/08.

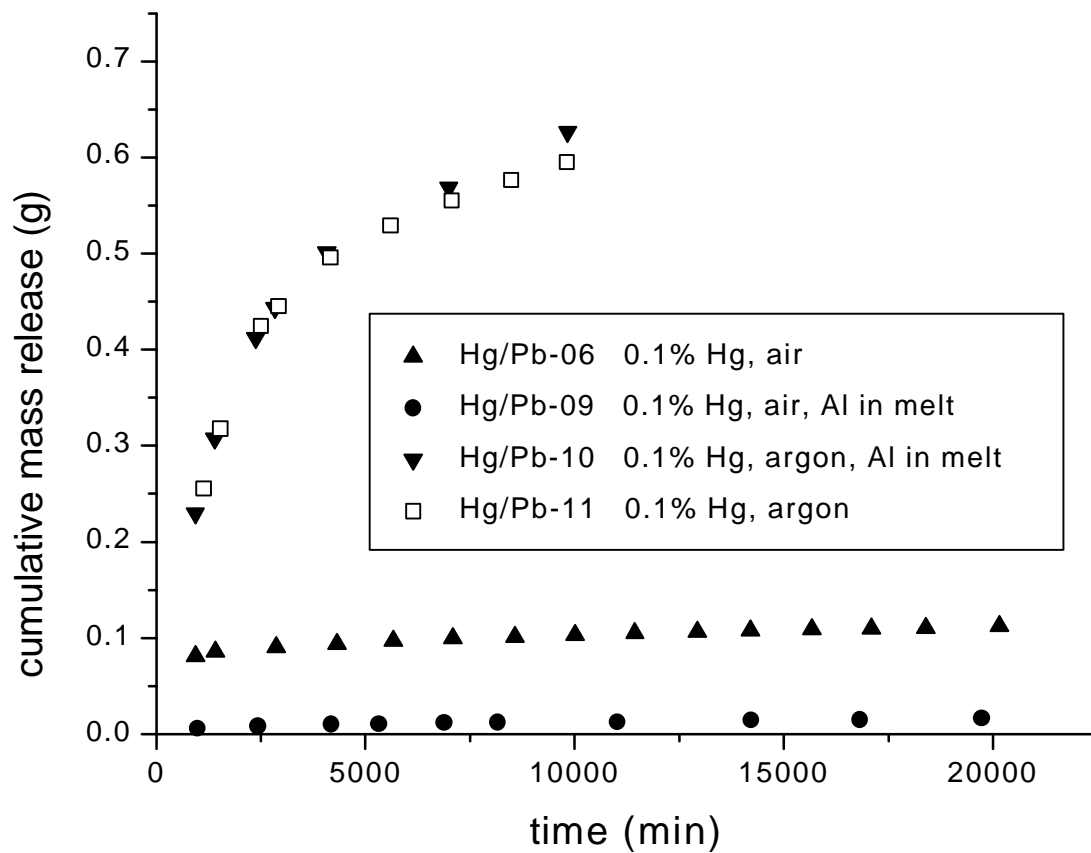


Figure 5. Comparison of the Cumulative Effects of Mercury Concentration, Atmosphere Composition and Aluminum in the Melt on the Rate of Vaporization of Elemental Mercury From Molten Lead Pools: Hg-Pb/06, Hg-Pb/09, Hg-Pb/10, Hg-Pb/11.

Table 1**Ranges of Nominal Test Parameters**

Parameter	Nominal Value and Uncertainty
Nominal Lead Mass	2.5 kg
Lead Surface Area	93.3 cm ²
Nominal Mercury Concentration (weight)	0.01%, 0.10%, 1.0%
Lead Temperature	340°C ± 2.5°C
Atmosphere Temperature	340°C ± 2.5°C
Gas Flow Rate	200 ml/min ± 6%, 750 ml/min ± 2%
Atmosphere Composition	argon (pre-purified, 99.998%), air
Melt Stirring Rate	none, 1 rpm
Aluminum in Melt	yes, no

Table 2**Specifications for Individual Tests**

Test (-)	Hg (wgt%)	Cover Gas	Gas Flow	Pool Agitation	Aluminum in Melt	Purpose/Comments
00	0.0	argon	low	no	no	Shakedown test, measure of the lead carryover.
01	1.0	argon	low	no	no	Effect of very high mercury concentration.
02	0.1	argon	low	no	no	Bounding (5-year APT) mercury concentration.
03	0.01	argon	low	no	no	Nominal (1-year APT) mercury concentration.
04	0.1	argon	high	no	no	Effects of changes in the gas flow rate.
05	0.1	argon	low	yes	no	Effects of agitation of the melt pool.
06	0.1	air	low	no	no	Effects of an oxidizing air atmosphere.
08	0.01	air	low	no	no	Effects of air at nominal mercury concentration.
09	0.1	air	low	no	yes	Effects of aluminum in the melt with air.
10	0.1	argon	low	no	yes	Effects of aluminum in the melt with argon.
11	0.1	argon	low	no	no	Repeat of Hg-Pb/02 with improved procedures.

Table 3**Cumulative Average Mercury Release Results**

Test (-)	Cover Gas	Initial Hg Mass	Mercury Concentration	Mercury Released	Duration of Test	Mercury RF	Release Rate, Average	Average Daily Release
01	argon	28.01 gm	1.14 %	1.310 gm	2.0 hr	4.7 %	6.99E-3 gm/cm ² ·hr	56.4 %
02	argon	2.83	0.11	0.330	48.0	11.7	8.02E-5	5.85
03	argon	0.31	0.01	0.031	165.0	10.2	2.19E-6	1.48
04	argon	2.79	0.11	0.155	50.4	5.6	3.57E-5	2.67
05*	argon	2.75	0.11	0.569	24.5	20.7	2.69E-4	20.3
06	air	2.58	0.10	0.112	336.0	4.4	3.86E-6	0.31
08	air	0.27	0.01	0.011	643.6	4.1	1.96E-7	0.15
09	air	2.51	0.10	0.017	328.9	0.7	5.82E-7	0.05
10	argon	2.41	0.09	0.626	163.9	26.0	4.43E-5	3.81
11	argon	2.55	0.10	0.596	163.6	23.4	4.22E-5	3.43

* Data shown are for the 24.5 hour period of melt agitation only. The time during which the melt was not being stirred has been excluded.

Table 4

Integrated Mercury Mass Released vs. Elapsed Time: Tests Hg-Pb/02 and Hg-Pb/03

Test Hg-Pb/02		Test Hg-Pb/03	
Elapsed Time (min)	Cumulative Release (g)	Elapsed Time (min)	Cumulative Release (g)
120	0.0444	310	0.0039
240	0.071	1290	0.0039
253	0.0787	1775	0.0039
480	0.1015	2685	0.0062
1140	0.1816	3155	0.0096
1595	0.21	9900	0.0312
2880	0.3321		

Table 5

Integrated Mercury Mass Released vs. Elapsed Time: Tests Hg-Pb/04 and Hg-Pb/05

Test Hg-Pb/04		Test Hg-Pb/05	
Elapsed Time (min)	Cumulative Release (g)	Elapsed Time (min)	Cumulative Release (g)
143	0.0085	595	0.0058
398	0.0398	835	0.0945
643	0.0561	1113	0.1848
1133	0.0757	1415	0.3357
1583	0.097	2065	0.5745
2048	0.1153	2575	0.633
2588	0.1312	4000	0.7358
2800	0.1398	5395	0.7464
3024	0.1553	6923	0.7678

Table 6**Integrated Mercury Mass Released vs. Elapsed Time: Tests Hg-Pb/06 and Hg-Pb/08**

Test Hg-Pb/06		Test Hg-Pb/08	
Elapsed Time (min)	Cumulative Release (g)	Elapsed Time (min)	Cumulative Release (g)
936	0.0811	2925	0.0024
1410	0.0856	5415	0.0036
2860	0.0906	8610	0.0060
4320	0.0939	21570	0.0082
5660	0.0972	30260	0.0097
7090	0.0996	38617	0.0109
8575	0.1009		
10015	0.1028		
11440	0.1049		
12940	0.1064		
14200	0.1078		
15671	0.1088		
17090	0.1097		
18390	0.1105		
20160	0.1123		

Table 7**Integrated Mercury Mass Released vs. Elapsed Time: Tests Hg-Pb/06 and Hg-Pb/09**

Test Hg-Pb/06		Test Hg-Pb/09	
Elapsed Time (min)	Cumulative Release (g)	Elapsed Time (min)	Cumulative Release (g)
936	0.0811	979	0.0057
1410	0.0856	2432	0.0081
2860	0.0906	4184	0.0104
4320	0.0939	5322	0.0107
5660	0.0972	6884	0.0119
7090	0.0996	8159	0.0123
8575	0.1009	11019	0.0126
10015	0.1028	14224	0.0146
11440	0.1049	16824	0.0149
12940	0.1064	19734	0.0165
14200	0.1078		
15671	0.1088		
17090	0.1097		
18390	0.1105		
20160	0.1123		

Table 8

Integrated Mercury Mass Released vs. Elapsed Time: Tests Hg-Pb/10 and Hg-Pb/11

Test Hg-Pb/10		Test Hg-Pb/11	
Elapsed Time (min)	Cumulative Release (g)	Elapsed Time (min)	Cumulative Release (g)
930	0.2296	1125	0.2558
1402	0.3076	1525	0.3179
2370	0.412	2495	0.4248
2835	0.443	2915	0.4454
4080	0.5015	4160	0.4962
6990	0.5683	5600	0.5294
9835	0.6264	7060	0.5552
		8480	0.5765
		9815	0.5955